Boltzmann theory of engineered anisotropic magnetoresistance in (Ga,Mn)As

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We report on a theoretical study of dc transport coefficients in (Ga,Mn)As diluted magnetic semiconductor ferromagnets that accounts for quasiparticle scattering from ionized Mn^{2+} acceptors with a local moment $S = 5/2$ and from non-magnetic compensating defects. In metallic samples Boltzmann transport theory with Golden rule scattering rates accounts for the principle trends of the measured difference between resistances for magnetizations parallel and perpendicular to the current. We predict that the sign and magnitude of the anisotropic magnetoresistance can be changed by strain engineering or by altering chemical composition.

In most (III,V) semiconductors, Mn^{2+} substitution on a cation (column III element) site introduces an $S = 5/2$ local moment and a valence band hole [1]. $Mn_xIII_{1-x}V$ diluted magnetic semiconductors [2–5] (DMSs) are ferromagnetic and metallic for Mn fractions larger than $x \sim 1\%$. Many magnetic properties of the most robustly ferromagnetic samples, those that have $\sim 5\%$ or more Mn and are annealed to reduce the density of compensating defects, appear to be adequately explained by virtual crystal approximation models in which disorder is ignored [6–9]. For example in bulk DMSs, this approach can account for ferromagnetic critical temperatures ∼100 K [7,10], the correlation between magneto-crystalline anisotropy and substrate lattice constant [7,8], the size and sign of the anomalous Hall effect [11], and several optical properties [7]. It has also been used to describe properties of DMS heterostructures [12–14] for which the simplifications afforded by neglect of disorder are particularly helpful.

In this letter we investigate theoretically the $T = 0$ dc-transport coefficients of (Ga,Mn)As ferromagnetic semiconductors. We find that relaxation-time-approximation solutions of the Boltzmann equation provide anisotropic magnetoresistance (AMR) estimates that are in good agreement with experiments [15,16]. Our results suggests that transport properties of these metallic ferromagnets can be understood within a conventional framework in which disorder is treated as a weak perturbation. We find that the conductivity varies by several percent when the magnetic order parameter is reoriented by a weak magnetic field, and predict that the magnitude and sense of this change depends on the chemical composition and on the substrate on which the thin film DMS ferromagnet is epitaxially grown. This spontaneous magnetoresistance anisotropy is the transport analog of magneto-crystalline anisotropy [7,8] which has approximately the same size relative to the total condensation energy of the ordered state. All results presented in this paper are for the (Ga,Mn)As DMS's. A large database that details our predictions for the AMR of many other host semiconductors over a wide range of compositions and strains is available on the internet [17].

We consider a microscopic Hamiltonian in which valence band holes interact with randomly located spins of substitutional Mn^{2+} impurities via exchange interactions, and with randomly located ionized defects and each other via Coulomb interactions. Focusing on $T = 0$, we assume that the Mn spins are fully aligned in the ferromagnetic ground state. In the virtual crystal approximation, the interactions are replaced by their spatial averages, so that the Coulomb interaction vanishes and hole quasiparticles interact with a spatially constant Zeeman field. The unperturbed Hamiltonian for the holes then reads $H_0 = H_L + J_{pd} N_{Mn^{2+}} S\hat{\Omega} \cdot \vec{s}$, where H_L is the six-band Kohn-Luttinger Hamiltonian [8], $\hat{\Omega}$ is the Mn local moment orientation, $J_{pd} = 55$ meV nm^3 [3] is the local-moment – valence-band-hole kinetic-exchange coupling constant, $N_{Mn^{2+}}$ is the density of ordered Mn local moments, and $\vec{s}$ is the envelope-function hole spin operator [8]. We use the relaxation-time-approximation solution to the semiclassical Boltzmann equation to estimate the dc conductivity tensor:

$$\sigma_{\alpha\beta} = \frac{e^2}{\hbar V} \sum_{n,k} (\hbar\Gamma_{n,\vec{k}})^{-1} \frac{\partial E_{n,\vec{k}}}{\partial k_\alpha} \frac{\partial E_{n,\vec{k}}}{\partial k_\beta} \delta(E_F - E_{n,\vec{k}}) \ , \quad (1)$$

where $\Gamma_{n,\vec{k}}$ is the quasiparticle elastic scattering rate, n and $\vec{k}$ are the band and wavevector indices of the valence band Bloch states of the unperturbed system, and $E_{n,\vec{k}}$ are the spin-split band energies of the ferromagnetic state. In Eq. (1) we have omitted the asymmetric terms in the off-diagonal elements of $\sigma_{\alpha\beta}$ that contribute to the anomalous Hall conductivity, discussed in detail elsewhere [11]. The symmetric off-diagonal elements, described by Eq. (1), vanish when the magnetization is aligned along one of the cube edges of the host lattice.

In our model, itinerant holes are scattered on substitutional Mn^{2+} impurities by a Thomas-Fermi screened Coulomb potential and by a magnetic-kinetic-exchange potential. For majority-spin holes both potentials are attractive while for minority-spin holes the magnetic potential becomes repulsive. We estimate the transport

weighted scattering rate from Mn^{2+} impurities using Fermi's golden rule:

$$\Gamma_{n,\vec{k}}^{Mn^{2+}} = \frac{2\pi}{\hbar} N_{Mn^{2+}} \sum_{n'} \int \frac{d\vec{k'}}{(2\pi)^3} |M_{n,n'}^{\vec{k},\vec{k'}}|^2 \times \delta(E_{n,\vec{k}} - E_{n'\vec{k'}})(1 - \cos\theta_{\vec{k},\vec{k'}}) \ , \quad (2)$$

where the scattering matrix element was approximated by the following expression,

$$M_{n,n'}^{\vec{k},\vec{k'}} = J_{pd} S \langle z_{n\vec{k}} | \hat{\Omega} \cdot \vec{s} | z_{n'\vec{k'}} \rangle - \frac{e^2}{\epsilon_{host}\epsilon_0(|\vec{k} - \vec{k}'|^2 + q_{TF}^2)} \langle z_{n\vec{k}} | z_{n'\vec{k'}} \rangle . \quad (3)$$

Here ϵ_{host} is the host semiconductor dielectric constant, $|z_{n\vec{k}}\rangle$ is a six-component envelope-function eigenspinor of the Hamiltonian H_0, and the Thomas-Fermi screening wavevector was approximated by the parabolic band expression, $q_{TF} = \sqrt{3e^2 p/(2\epsilon_{host}\epsilon_0 E_F)}$, where p is the itinerant hole density and E_F is the Fermi energy.

Recent experiments have established that magnetic and transport properties of (III,Mn)V DMS ferromagnets are sensitive to post-growth annealing protocols [4,5,15,18], and that this sensitivity is associated with changes in the density of defects that compensate the Mn^{2+} acceptors. Our model recognizes that the transport properties of these materials are not determined solely by the scattering from substitutional Mn^{2+} impurities and allows explicitly for scattering from compensating defects. We assume that compensation can occur due to the presence of As-antisite defects (common in low-temperature MBE grown GaAs hosts) or due to Mn interstitials. As-antisite defects are non-magnetic and contribute only $Z = 2$ Coulomb scattering. Mn interstitials, when they are present [18,16], are unlikely to be magnetically ordered and can also be modeled as $Z = 2$ donors [19]. Overall charge neutrality implies that the density of holes is $p = N_{Mn^{2+}} - 2N_c$, where N_c is the density of compensating impurities.

Rough estimates for the $T = 0$ quasiparticle scattering rates can be obtained from parabolic-band-approximation expressions for majority heavy-hole states assuming Mn^{2+} kinetic-exchange scattering only, $\Gamma_{pd} = (N_{Mn^{2+}}) J_{pd}^2 S^2 m^* \sqrt{2m^* E_F}/(4\pi\hbar^4)$, or Mn^{2+} and As-antisite Coulomb scattering only which leads to scattering rate Γ_C given by the Brooks-Herring formula [20]. (We show below that most of the current in a (Ga,Mn)As ferromagnet is carried by majority spin heavy-holes.) Taking the heavy-hole effective mass $m^* = 0.5 m_e$, $p = 0.4$ nm^{-3} and Mn fraction $x = 5\%$, we obtain $\hbar\Gamma_{pd} \sim 20$ meV and $\hbar\Gamma_C \sim 150$ meV. Our full numerical six-band calculations are consistent with these estimates, and in particular predict that the Coulomb contribution to the elastic scattering rate is several times larger than the kinetic-exchange contribution for typical chemical compositions. The total scattering rate, averaged over the majority heavy-hole Fermi surface, decreases with increasing density of itinerant holes for a fixed Mn^{2+} concentration. One important conclusion of this analysis is that even in the heavily doped and compensated (Ga,Mn)As DMSs, the lifetime broadening of the quasiparticle ($\hbar\Gamma$) is smaller than the valence band spin-orbit coupling strength ($\Delta_{so} = 341$ meV), which partially justifies the neglect of disorder [11] in evaluating some properties, *e.g.* the anomalous Hall conductivity.

In Fig.1 we plot σ_{xx}, calculated numerically using the six-band Kohn-Luttinger model and Eqs. (1) and (2), for a fully strained $Mn_{0.06}Ga_{0.94}As$ sample grown on a GaAs substrate and assuming compensation due to As-antisites alone (i.e. no Mn-interstitials present). The substrate – DMS lattice mismatch, $e_0 \equiv (a_{sub} - a_{DMS})/a_{DMS}$, is between -0.002 and -0.003 in this case [2,16]. (Note that a_{sub} and a_{DMS} are the lattice constants of a fully relaxed substrate and ferromagnetic layer, respectively.) The parameters of the six-band Kohn-Luttinger model and strain coefficients used in these calculations are given in [21]. We also show in Fig.1 separate contributions from individual heavy- and light-hole bands and demonstrate that in the ferromagnetic state the current is carried mostly by the majority-spin heavy-holes, a property that will be important for understanding the spin-injection properties of (III,Mn)V DMS ferromagnets. The absolute conductivities predicted by our model are reasonably consistent with experiment [4,5,15]. For lower Mn concentrations ($x < 4\%$) the theoretical conductivities become several times larger than the measured values due, we believe, to some combination of inaccuracy in our scattering amplitude estimates, sources of disorder in current experimental samples that we do not account for in the model, and especially at small x coherent scattering effects that eventually lead to localization.

As mentioned above, strong spin-orbit coupling in the semiconductor valence band leads to a variety of magneto-anisotropy effects [7,8,22]. For dc transport the in-plane conductivity along, *e.g.* the x-direction should change when the magnetization is rotated by applying a magnetic field stronger than the sample's magneto-crystalline anisotropy field. In Fig. 2 we show the anisotropic magnetoresistance coefficients, $AMR_{ip} \equiv [\rho_{xx}(\hat{\Omega}||x) - \rho_{xx}(\hat{\Omega}||y)]/\rho_{xx}(\hat{\Omega}||y)$ and $AMR_{op} \equiv [\rho_{xx}(\hat{\Omega}||x) - \rho_{xx}(\hat{\Omega}||z)]/\rho_{xx}(\hat{\Omega}||z)$, for orthogonal magnetization directions in the plane of the thin ferromagnetic layer, and for one of the magnetization directions along the growth direction, respectively. The Mn fraction assuming As-antisite compensation alone is indicated in the figure by x_1. The total Mn fraction (including substitutional Mn^{2+} atoms and Mn-interstitial atoms) for compensation due to interstitial Mn alone is labeled by x_2. The plots demonstrate that in (Ga,Mn)As/GaAs ferromagnets, which have compressive strain ($e_0 < 0$), AMR is negative for typical chemi-

cal compositions, and $|AMR_{ip}| < |AMR_{op}|$. (Note that $AMR_{ip} = AMR_{op}$ in unstrained cubic DMSs.) The main plot shows that for a fixed hole density ($p = 0.4$ nm^{-3}) the magnitude of the AMR decreases from $\sim 10\%$ to $\sim 1\%$ with increasing Mn fraction. All those observations are consistent with available experimental data on (Ga,Mn)As DMS's [15,16]. A more detailed comparison to measurements by Gallagher *et al.* [16] shows that theoretical data assuming As-antisite compensation only overestimate the decrease of AMR with increasing Mn fraction. Better quantitative agreement is obtained for compensation from Mn-interstitials, which is consistent with the presumed dominance of Mn-interstitial defects over As-antisite defects in the samples measured by Gallagher *et al.* [16].

In addition to this AMR engineering through doping that has been confirmed experimentally, our theory predicts a large sensitivity of the spontaneous transport anisotropy to strain. While strain does not play a significant role for AMR_{ip}, as one might expect from symmetry considerations, AMR_{op} can change by more than 10% over the range of strains that can be achieved in the thin ferromagnetic layers by a proper choice of the substrate [7]. For larger Mn concentrations, AMR_{op} is predicted to become positive in samples with tensile strain, as shown in the main plot of Fig. 2.

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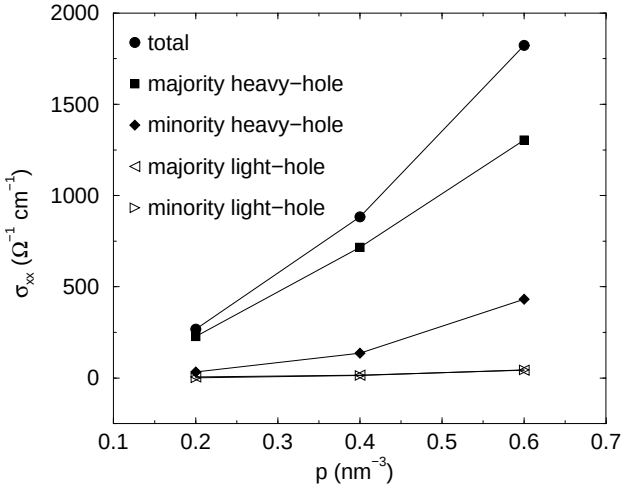

FIG. 1. Conductivity for measuring current and magnetization directed along the x-axis in the plane of the (Ga,Mn)As film as a function of the total hole density. These results were obtained for a GaAs semiconductor host doped with 6% Mn and with strain $e_0 = -0.002$. No Mn-interstitial atoms are assumed to be present in the ferromagnetic layer. For typical hole densities the current is carried mostly by the majority heavy-holes.

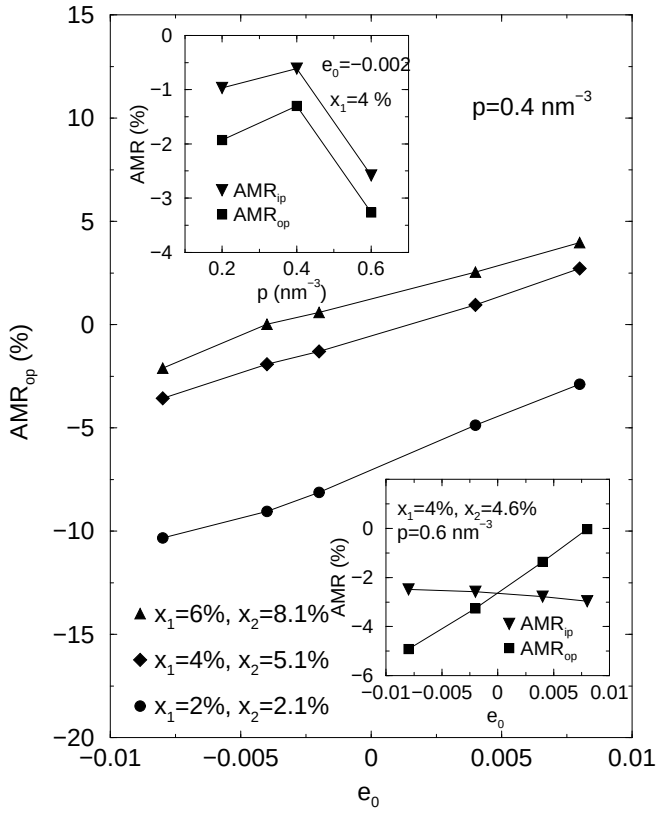

FIG. 2. Anisotropic magnetoresistance coefficients as a function of strain (main plot and lower inset) and hole density (upper inset). Mn fractions corresponding to compensation due to As-antisites alone and Mn-interstitials alone are labeled as x_1 and x_2 respectively.